A regional application of the MAGIC model in Wales: calibration and assessment of future recovery using a Monte-Carlo approach

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Abstract

A survey and resurvey of 77 headwater streams in Wales provides an opportunity for assessing changes in streamwater chemistry in the region. The Model of Acidification of Groundwater In Catchments (MAGIC) has been calibrated to the second of two surveys, taken in 1994–1995, using a Monte-Carlo methodology. The first survey, 1983–1984, provides a basis for model validation. The model simulates a significant decline of water quality across the region since industrialisation. Agreed reductions in sulphur (S) emissions in Europe in accordance with the Second S Protocol will result in a 49% reduction of S deposition across Wales from 1996 to 2010. In response to these reductions, the proportion of streams in the region with mean annual acid neutralising capacity (ANC) >0 is predicted to increase from 81% in 1995 to 90% by 2030. The greatest recovery between 1984 and 1995 and into the future is at those streams with low ANC. In order to ensure that streams in the most heavily acidified areas of Wales recover to ANC zero by 2030, a reduction of S deposition of 80–85% will be required.

Introduction

Large areas of Wales are vulnerable to acidification, especially the uplands, as the bedrock is slow weathering and the soils have little or no acid neutralising capacity (Hornung *et al.*, 1990). It is estimated that 34% of soils in Wales are affected by acidic deposition and in these areas, about 50% of the first to third order streams may have been damaged. In terms of nature conservation, Wales is the worst affected region in the UK with more than 40% of the total area of Sites of Special Scientific Interest (SSSIs) potentially damaged by freshwater acidification (Stevens *et al.*, 1997). Against this background and given a 52% reduction in UK sulphur dioxide emissions between 1980 and 1995, stream chemistry surveys have been conducted across Wales during 1983–84 and 1995–96 to determine the degree of chemical change.

Dynamic models provide the only opportunity to determine the likely future response of soil and stream chemistry to changes in deposition of S with an indication of the time over which recovery will be achieved in response to reduced acidic oxide emissions. MAGIC (Model for Acidification of Groundwater In Catchments) is a dynamic, lumped, process-based model of soil and water chemistry which represents the processes of major importance to soil acidification and is driven by trends in S deposition (Cosby *et al.*, 1985a,b). It has been extensively applied to sites in America and Europe (Macmillan and Ferrier 1994; Whitehead *et al.*, 1993; Waters and Jenkins 1992; Forti *et al.*, 1994). Recently, the model has been applied to the Acid Waters Monitoring Network (AWMN), a set of 22 catchments located in the acid sensitive areas of the UK (Jenkins *et al.*, 1997; Helliwell *et al.*, 1998). The model has been tested against laboratory experiments, land use change and whole catchment manipulation experiments (Wright *et al.*, 1994; Cosby *et al.*, 1995).

For regional application of MAGIC, two 'scaling up' methods have previously been used. The first is a multiple-site approach where numerous catchments in a region are calibrated and impacts may be assessed for a particular location or for the region as a whole. The disadvantages of such an approach are the heavy data requirements and the fact that impacts at streams not covered by the survey cannot be readily inferred. To date, this methodology has been applied in Norway (Frogner *et al.*, 1994; Cosby and Wright, 1998), the U.S. (Turner *et al.*, 1992) and in the Galloway region of Scotland (Wright *et al.*, 1994). The second method uses a Monte-Carlo approach whereby input variables are randomly sampled from defined frequency distributions based on observed data describing the whole region for multiple runs, or 'realisations', of the

model. This approach, which has been applied in Norway (Hornberger *et al.*, 1989; Cosby and Wright, 1998) and Wales (Jenkins *et al.*, 1990), is limited in that impacts cannot be quantified for a given location within the region. However, this is traded for its strengths in supporting policy decisions by estimating statistics for the future behaviour of surface waters in the region as a whole, such as the percentage of streams likely to recover under a given deposition scenario. A comparison of the two approaches has been made in Norway (Cosby and Wright, 1998) and Wales (Collins and Jenkins, 1998).

A Monte-Carlo regional methodology, based on that used for a calibration to the first Acid Waters survey (Jenkins *et al.*, 1990) has been used to make predictions of future surface water chemistry in Wales. The first survey of these sites was carried out during 1984. The regional application provides a simulation of the changes in the frequency distribution of streams across Wales from 1984 to 1995 and predicts the changes from 1995 to 2010 and 2030. Long-term monitoring at the 4 Welsh catchments in the Acid Waters Monitoring Network (Patrick *et al.*, 1995) provides a context for the two survey years but tells little about trends in surface water or deposition chemistry over the intervening time period. Here, MAGIC is applied using a Monte-Carlo regional methodology to test the usefulness of the model for simulation of the response to acidification across Wales and, furthermore, to evaluate the effects of future reductions in sulphur (S) emissions on recovery of surface water acidity.

Survey region

The study area lies in western Wales (Fig. 1), a region characterised by an inland plateau between 200 and 600 m in altitude with the mountainous Snowdonia area in the northwest. Surveys of wet deposition in Wales show deposition of non-marine sulphate (SO_4) and nitrate (NO_3) related mainly to rainfall amount, and hence topography (Reynolds and Ormerod, 1993). In general, concentrations of non-marine SO_4 and NO_3 in bulk deposition are relatively low compared with more polluted areas of Europe, but fluxes are high because rainfall exceeds 2 m yr^{-1} in the mountains. In 1994, for example, wet deposition exceeded 10 kg S ha^{-1} yr^{-1} and 5 kg N ha^{-1} yr^{-1} at high altitude (Vincent *et al.*, 1995). The study area is underlain by rocks of Cambrian, Ordovician and Silurian age, and soils are thin and base poor in the uplands. The combination of high acid deposition fluxes and vulnerable geology gives the region its documented low critical loads and high exceedance compared to much of the UK (Battarbee, 1995).

The regional bulk deposition concentrations for 1995 (Table 1) are the means of the 19 upland sites in the survey. Dry deposition of Cl and SO_4 is added to this such that input equals output. Dry deposition of base cations is then assumed to be proportional to Cl and the ionic ratios

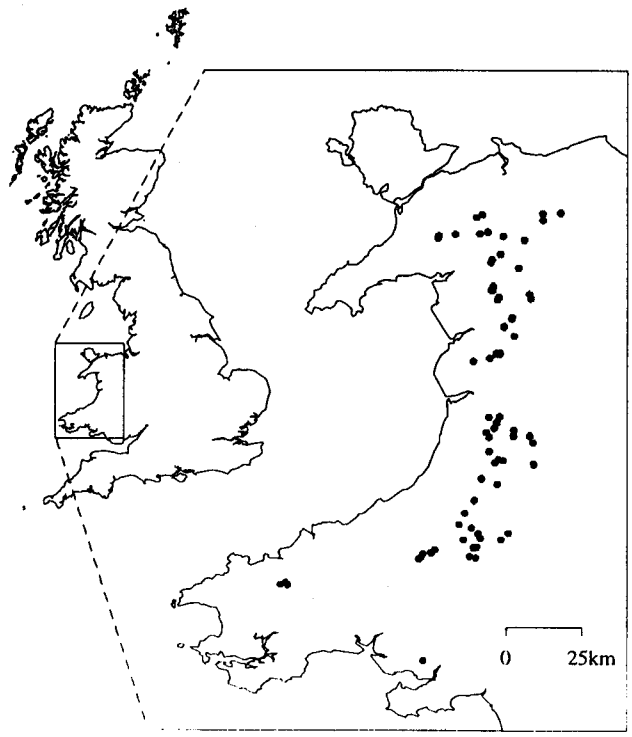

Fig. 1. *Location of 77 stream sampling points common to both Welsh Acid Waters surveys (1984 and 1995).*

in seawater. This extra flux of ions is assumed to incorporate dry deposition and enhanced deposition in forests.

For N, the total deposition of reduced and oxidised N, as modelled by the Institute of Terrestrial Ecology at Bush, were used (Fowler, pers. comm.). N deposition is assumed to have increased since industrialisation to 1984 in accordance with estimated NO_3 emissions in the UK (DOE, 1983, 1990) and then held constant to 1995 (Fig. 2). The historical trend in wet-deposited non-marine SO_4 (Fig. 2) is assumed to follow the sequence described by the Warren Spring Laboratory (1983, 1987, unpublished data). The decline from 1984 to 1995 of 19.3% is calculated from observations at Acid Deposition Network sites in Wales.

Table 1. Means for 19 sites of 1995 observed bulk deposition (μeq m^{-2} yr^{-1}) from the Acid Deposition survey and estimated total deposition.

	Acid Waters wet deposition measurement	Total deposition
Ca	9.5	12.7
Mg	23.0	39.3
Na	92.6	165.7
K	2.8	2.6
NH_4	21.8	55.7
SO_4	40.5	93.7
Cl	117.2	202.4
NO_3	16.1	21.9

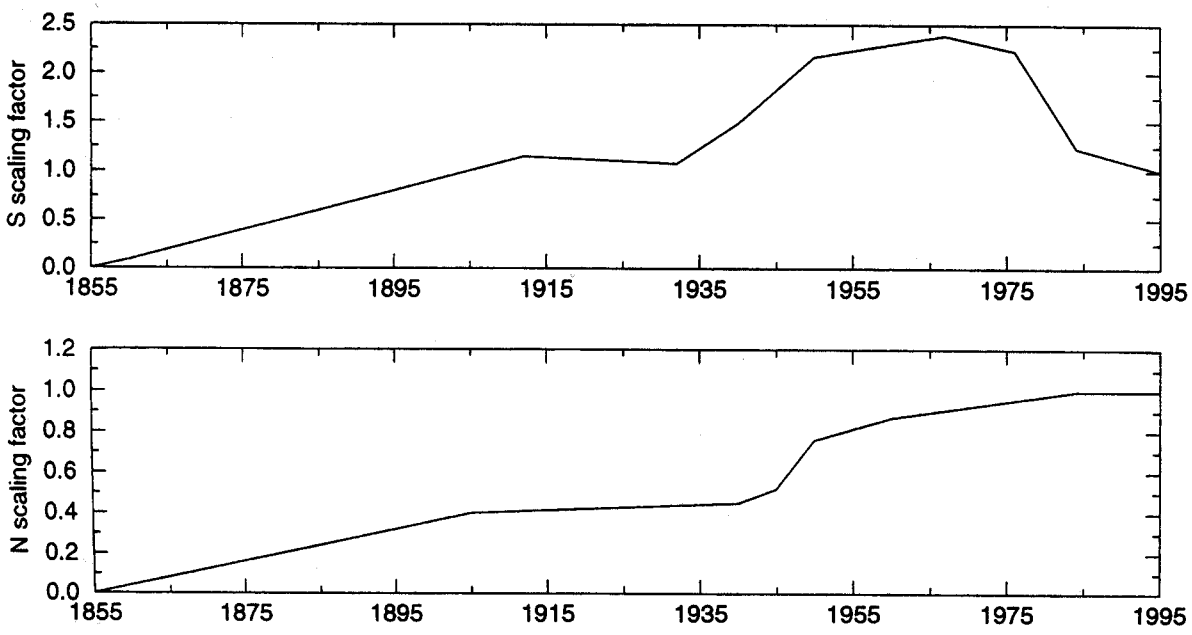

Fig. 2. *Estimated historical trend in S and N deposition relative to present-day (1995) deposition flux.*

This compares with a 15% reduction in wet non-marine S deposition across the UK from 1986 to 1994 with no corresponding change evident in the NO_3 or ammonia (NH_4) deposition (Vincent *et al.*, 1995). The lower stream concentration of Cl in 1995 relative to 1984 (Tables 2 and 3) is modelled by assuming decreased deposition of sea salt.

The frequency distribution for runoff was derived from 30 gauged streams at locations adjacent to the 1984 Welsh Acid Waters deposition monitoring sites. The distribution of runoff has a mean value of 1.55 m with minimum and maximum of 0.50 m and 3.17 m, respectively. Rainfall is inferred by assuming evapotranspiration of 15% across the region, reflecting reported values of 10% for moorland and 20% for forests in the region (Jenkins *et al.*, 1997). There is no attempt to model annual variation or longer-term trends in runoff.

In all 130 streams were sampled in the 1984 survey and 120 in the 1995 survey. Of these 77 streams are in common and used in this analysis. The pH of streams in 1984 varied from 4.9 to 6.9 and increased slightly between the two surveys; the mean rose from 6.1 to 6.3 (Tables 2 and 3). Significantly decreased conductivity, K, SO_4 and Cl are also observed (Tables 2 and 3). A full analysis of the observed differences in chemistry is presented by Stevens *et al.* (1997).

The long-term context for the changes observed between the two survey years is provided by data from the Acid Waters Monitoring Network (Patrick *et al.*, 1995). Three Welsh sites in this network have been monitored since 1988; Llyn Llagi and Llyn Cwm Mynach, in north west Wales, and Afon Hafren at Plynlimon in central Wales. At all three, alkalinity decreased from 1988 to 1993

Table 2. Annual geometric mean stream chemistry (μeq l^{-1}) for the 1984 Acid Waters survey in Wales. Only the sites in common with the 1995 survey are included.

1984	N	Mean	Standard deviation	Minimum	Maximum
pH	77	6.1	0.5	4.8	6.9
Ca	77	106	45	38	238
Mg	77	96	30	45	200
Na	77	212	46	148	367
K	77	9	5	4	43
SO_4	77	137	48	44	280
Cl	77	263	63	182	481
NO_3	77	19	15	4	73

Table 3. Annual geometric mean stream chemistry (μeq l^{-1}) for 1995 for the 1995 Acid Waters survey in Wales. Only the sites in common with the 1984 survey are included.

1994–95	N	Mean	Standard deviation	Minimum	Maximum
pH	77	6.2	0.5	4.9	6.9
Ca	77	116	64	36	321
Mg	77	98	37	43	227
Na	77	216	55	146	379
K	77	6	5	2	38
SO_4	77	113	47	51	294
Cl	77	240	65	151	427
NO_3	77	17	18	2	86

and at two of the three, pH also decreased. This continuing acidification contrasts with the survey results which suggest a recovery across the region. Of the 77 surveyed sites, however, 18 suffered a decline in pH between the two surveys despite the mean recovery across the region as a whole.

Ranges for soil parameters (depth, bulk density, cation exchange capacity and present-day exchangeable fractions of base cations) used in the model (Table 4), are derived from empirical data and literature. SO_4 adsorption is assumed negligible, the aluminium (Al) equilibrium constant in the soil (KAl) is set at 9.2, and the partial pressure of carbon dioxide in the soil (pCO_2) at 20 times atmospheric. The mean concentration of organic acid in soil solution was set at 96 mmol m^{-3}, and dissociation constants were set at 4.5, 8.0 and 16.0 for the monoprotic, diprotic and triprotic organic acids, respectively. Mean annual temperature is assumed to be 5°C in soil and stream waters.

Regional application of the MAGIC model

The use of a Monte-Carlo technique enables the incorporation of variability into the model to account for heterogeneity in physical and chemical characteristics of soil and water across the region and uncertainty in measurement of physical and chemical parameters. Frequency distributions are defined for 16 parameters: runoff, streamwater concentrations of chloride (Cl), SO_4 and nitrate (NO_3), total deposition of reduced N, soil depth, soil bulk density and weathering rates and initial exchangeable fractions for base cations calcium (Ca), magnesium (Mg), sodium (Na) and potassium (K), and cation exchange capacity (CEC). Where spatial uniformity is assumed, parameters remain fixed across the region.

Frequency distributions for spatially-variable parameters are based on observed deposition, stream and soils data across the region. For each model realisation, input parameters are randomly sampled from these distributions to simulate present-day observed distributions of stream chemistry (Jenkins *et al.*, 1990). It is not possible to validate the historical simulation regionally in the way that site simulations have been validated by pH reconstructions from diatom fossil assemblages in lake sediment cores (Jenkins *et al.*, 1997). Calibrations are assumed to be satisfactory if simulated agree with observed present-day stream and soil chemistry. Target windows are defined for stream and soil chemistry determinands, using the 1995 data (Table 3).

Forecasts to 2030 are driven by mean modelled reductions in S deposition across the region. In addition, best and worst case scenarios are applied giving a band of uncertainty for the region as a whole. Future N deposition is assumed to remain constant at present-day levels.

The inputs of rainfall, weathering rates, initial exchangeable fractions of base cations and soil depth, bulk density and cation exchange capacity (CEC) are described by frequency distributions for the region (Table 4) and were sampled randomly for each of 5000 realisations. The model was calibrated to frequency distributions of 77 streams in the 1995 survey data. A total of 4191 simulations of the 5000 realisations were accepted.

In general, the parameter space is well covered by the accepted simulations (Table 4) with the simulated ranges of each of the 12 parameters covering at least 99% of the input range, and none covering less than 79% of the input range. Similarly, all average values, which are modes for triangular and means for normal distributions, are within 12% of the observed average. Calibrated weathering rates and soil exchange selectivity coefficients compare closely with those reported for the earlier regional MAGIC application to the 1984 survey (Jenkins *et al.*, 1990) (Table 5)

Calibration and validation

Observed and simulated stream chemistry for 1995 (Figs. 3, 4 and 5) demonstrate a satisfactory calibration for ANC

Table 4. Ranges of parameters used in the Monte-Carlo simulations (n = 5000) and resulting accepted simulations (n = 4191).

		Monte-Carlo			Accepted simulations		
	units	average	min	max	average	min	max
Stream parameters							
Qs	m	1.5	0.5	3.2	1.6	0.7	3.0
Cl[a]	meq l^{-1}	240	151	427	238	164	410
excess SO_4[a]	meq l^{-1}	88	6	267	85	20	227
NO_3[a]	meq l^{-1}	17	2	86	15	2	85
Soil parameters							
Depth	m		0.5	1.5		0.5	1.5
BD	kg m^{-3}		60	1590		61	1590
CEC	meq kg^{-1}		20	520		20	520
WeCa	meq m^{-2} yr^{-1}		5	200		5	200
WeMg	meq m^{-2} yr^{-1}		5	60		5	60
WeNa	meq m^{-2} yr^{-1}		0	30		0	30
WeK	meq m^{-2} yr^{-1}		0	5		0	5
ECa			0.5	45		0	45
EMg			0.5	35		0.5	35
ENa			0.5	15		0.5	15
EK			0.1	5		0.1	5
Deposition parameter							
NH_x-N dep	meq m^{-2} yr^{-1}	94	68	126	98	71	124

Notes: [a] observed
WeCa is weathering of Ca
ECa is exchangeable fraction of Ca (% of CEC)

and pH. The quality of the calibration of those determinands assumed to be conservative is governed by the suitability of the input distribution used to define the observed data and any bias in the distribution of acceptable model runs. Stream SO_4 concentrations are a good approximation to a normal distribution, although slightly positively-skewed towards high concentrations. Cl is also best represented as a normal distribution, and is similarly poor at the high extreme (Fig. 3d).

The cations (Fig. 4) have been calibrated by adjusting the input distributions of 8 soil chemistry Monte-Carlo parameters; the base cation weathering rates and exchangeable fractions. The K error appears larger than those of other base cations, but is small in terms of equivalents since concentrations are much lower. High cation concentrations are often underestimated, presumably when they coincide with high anion loads which are also underestimated.

Table 5. Calibrated regional soil selectivity coefficients (log_{10}) and weathering rates (meq m^{-2} yr^{-1}) for 4191 acceptable model simulations.

	mean	standard deviation	minimum	maximum
Ca/Al selectivity	1.53	3.07	–5.21	11.39
Mg/Al selectivity	1.75	2.61	–4.05	11.68
Na/Al selectivity	–1.09	1.52	–4.35	5.29
K/Al selectivity	–4.54	1.66	–8.31	1.72
Ca weathering	109	53	5	200
Mg weathering	33	16	5	60
Na weathering	15	9	0	30
K weathering	2	1	0	50
Total weathering	159	56	24	289

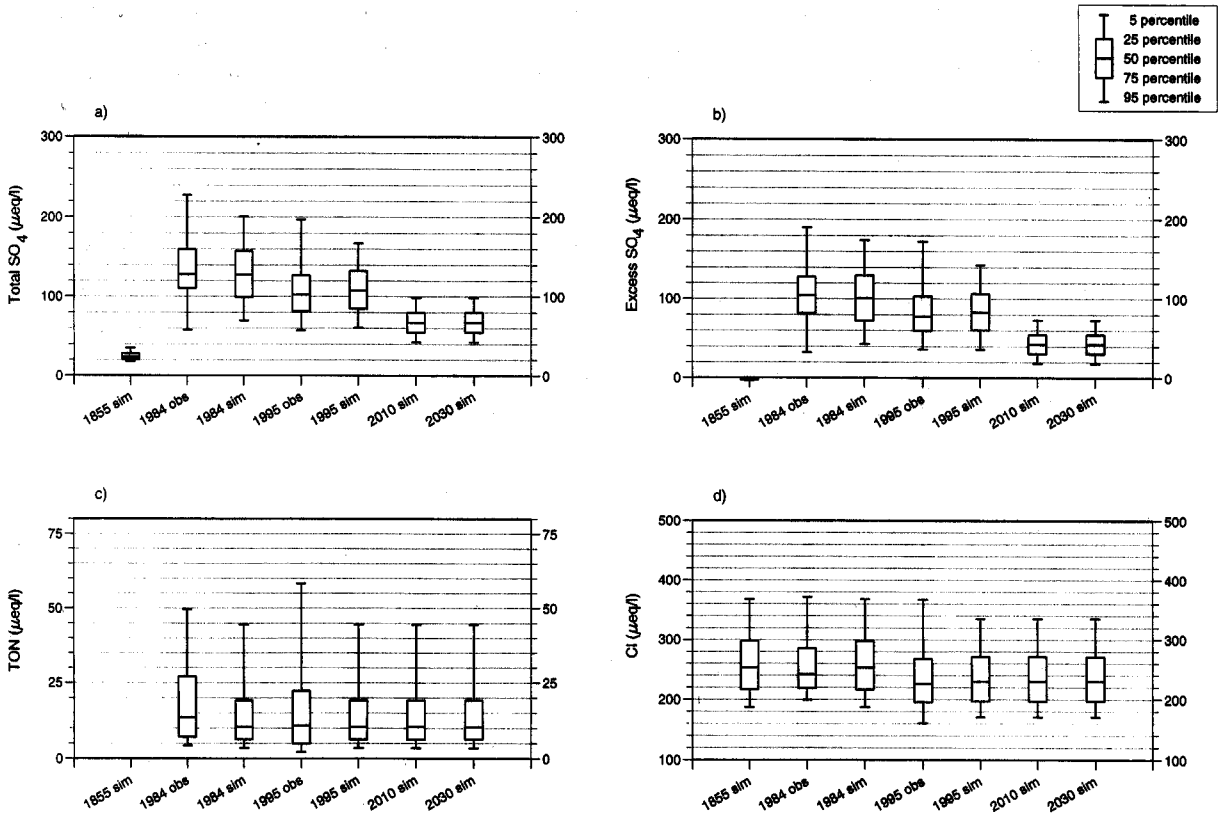

Fig. 3. *Bar and whisker plots of observed and simulated regional frequency distributions of anions in streamwater for 1984 and 1995.*

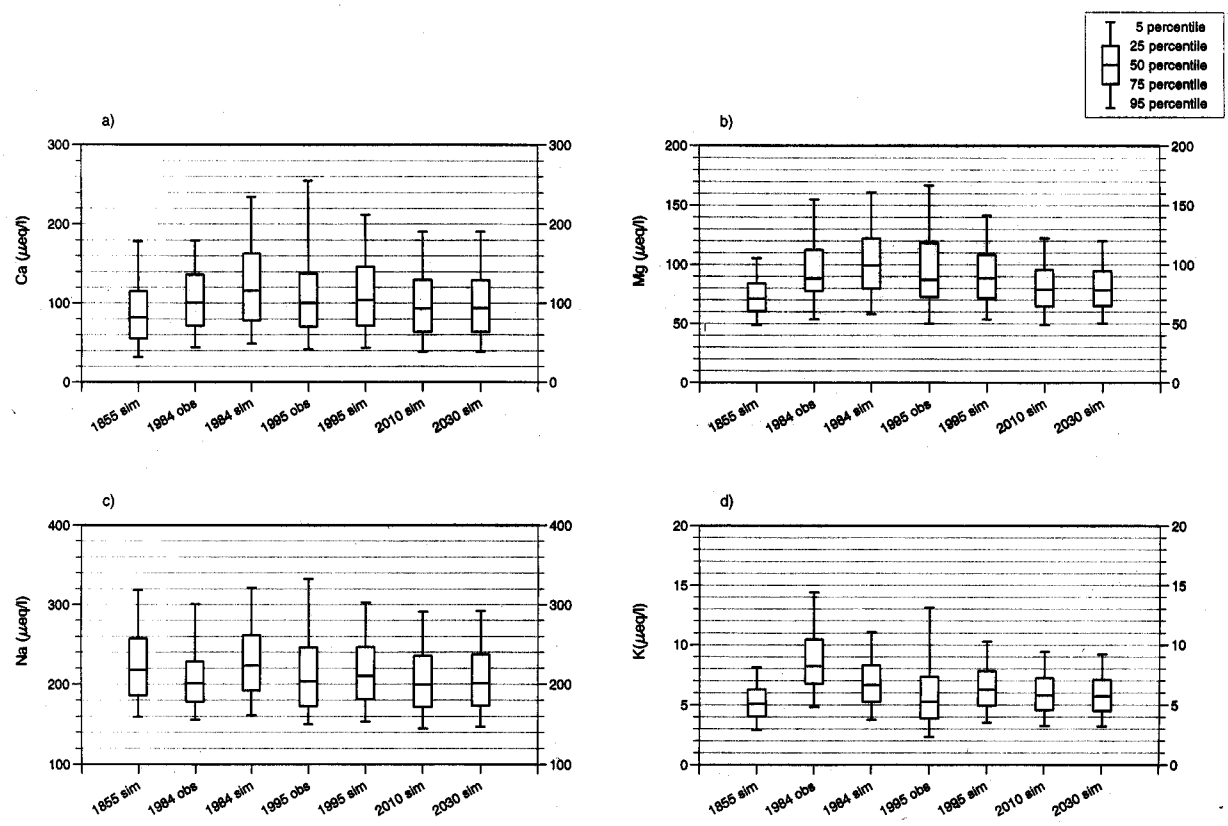

Fig. 4. *Bar and whisker plots of observed and simulated regional frequency distributions of base cations in streamwater for 1984 and 1995.*

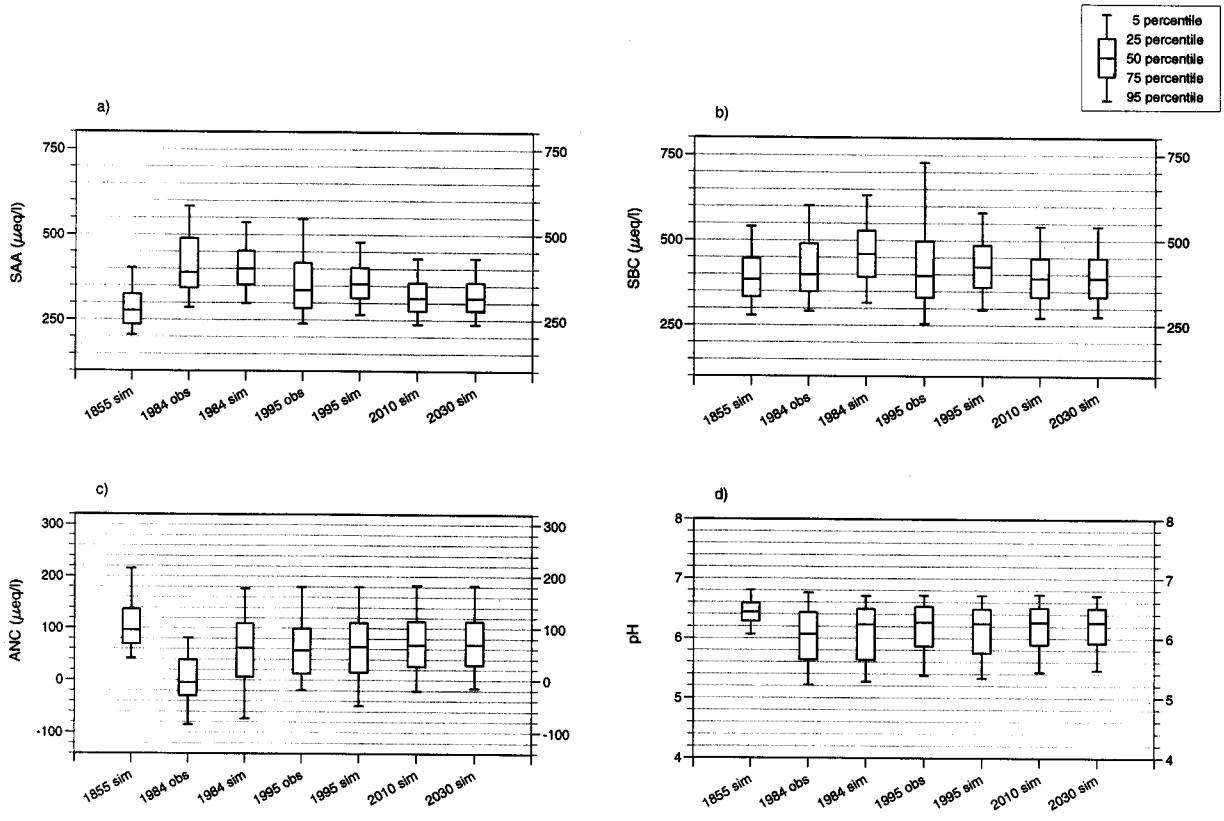

Fig. 5. *Bar and whisker plots of observed and simulated regional frequency distributions of sum of strong acid anions (SAA), sum of base cations (SBC), ANC and pH in streamwater for 1984 and 1995.*

The quality of the pH calibration (Fig. 5d) is achieved at the expense of precise Al calibration because of the regionalisation methodology. In the model simulations, Al buffers the low pH sites, and bicarbonate buffers the high pH sites. In reality, intermediate pH is buffered by the combination of cations as determined by the fixed parameters such as the solubility of Al, the partial pressure of CO_2 and the concentration of organics in the stream. At a regional scale, these parameters are fixed, and the model is not always able to balance these processes to simulate observed pH resulting in underestimation of the number of sites with pH in the range 5.3 to 6.0 and a bimodal distribution results. To reduce high pH, the pCO_2 in the stream was increased from 2 times atmospheric pressure to 4.5 times atmospheric pressure, reducing the amount of degassing and suppressing pH. Analysis of both surveys indicated a pCO_2 of between 3 and 6 times atmospheric pressure. Underestimated low pH was elevated by 2 mechanisms. First, surface water KAL was increased to 11.0 to lower the concentration of H^+ relative to Al^{3+}. Secondly, the organics in the surface water were set at 30 mmol m^{-3}, and the pH at which 50% of monoprotic organics dissociate at 6.5. The effect on Al was overestimation of the variance such that too many realisations had concentrations <1 μeq l^{-1} (41% compared with 2% of observations) and 12% had concentrations in excess of the observed maximum of 33 μeq l^{-1}.

Once calibration to 1995 stream chemistry is achieved, the hindcast reconstruction enables comparison of observed and simulated data for 1984. Where there is confidence in the measurement of determinands, this technique provides validation of the model. Conversely, confidence in the model has the potential to identify poor quality data. Here, comparison of the reconstructed data with observed data for 1984 provides validation of the model, but this becomes compromised by a question of quality surrounding the 1984 dataset. Relative to 1984, anions in 1995 have decreased but there is no corresponding decrease in cations, except K. In fact, the number of observations with high Ca, Na and Al has increased. It is implausible that a model based on mobile anion theory such as MAGIC, will simulate this change in cations, given a decrease in anions.

The hindcast simulation to 1984 confirms this. Whilst the direction and magnitude of change simulated for the anions is, broadly speaking, correlated with observations, each of the base cations in the simulation decreases. This occurs for each realisation (Fig. 6). The changes in SO_4 deposition are transferred directly to the stream as decreases in strong acid anions (SAA). A similar effect is apparent for the base cations (SBC), where the greatest response is at those sites which are strongly buffered. Recovery in ANC occurs at many sites as a consequence

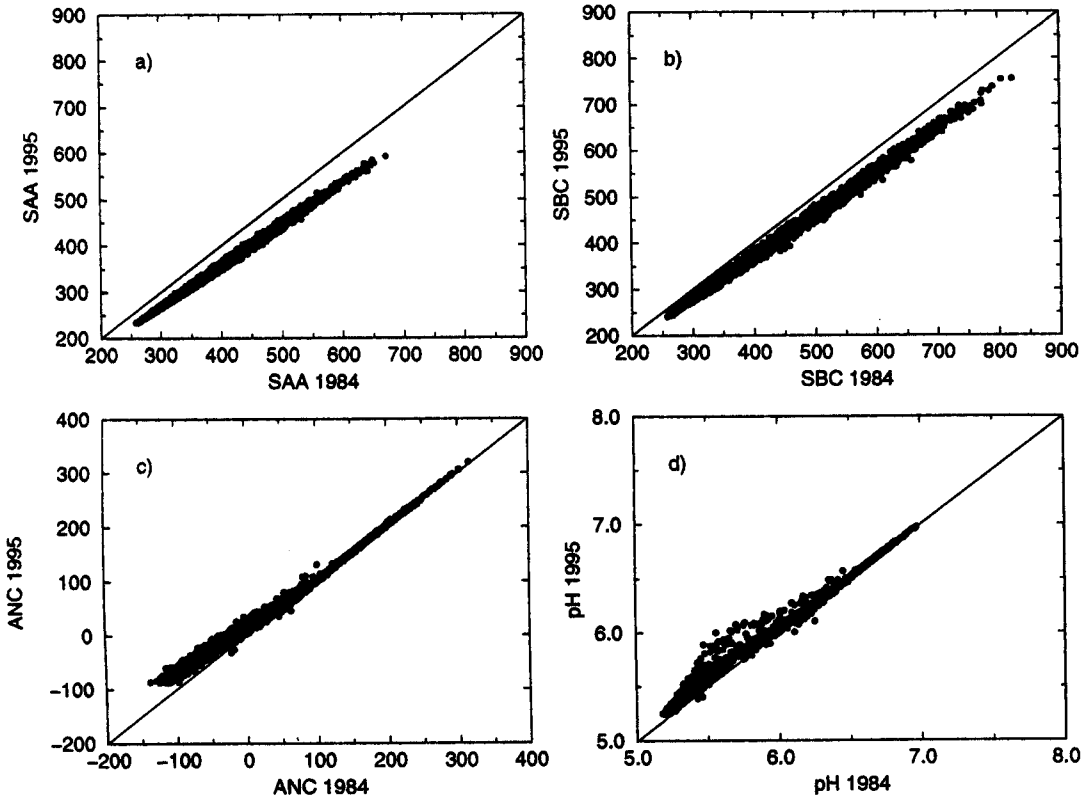

Fig. 6. *Scatter plots of simulated SAA, SBC, ANC and pH in 1984 and 1995 for 4191 acceptable simulations.*

of the decreased anions, and is greatest under most acidified conditions. Although the direction of change is the same in observed and simulated data, it is greater in the former because there is no change in observed SBC concomitant with the observed change in SAA.

Predicted recovery under the second sulphur protocol

The Second S protocol signed in June 1994 has committed the UK to reducing national S emissions by 70% by the year 2005 and by 80% by the year 2010. The HARM model (Metcalfe and Wyatt, 1995) is a source-receptor deposition model which is driven by a simple representation of meteorology and provides estimates of S deposition at a 20 km × 20 km resolution. Using HARM modelled present-day, non-marine S deposition (1989–1992) and predicted deposition for the years 2005 and 2010 a forecast sequence has been derived based on means of all HARM grid cells in Wales. The reductions were rescaled to the first year of the forecast such that, for example, the HARM reductions from 1996 become 38% and 49% of present day to 2005 and 2010, respectively.

Uncertainty in the HARM model is represented by two scenarios, the best and worst cases derived using a Monte-Carlo technique on the model parameters. Previous studies have found that at the site scale, uncertainty in the HARM model was significantly smaller than uncertainty incorporated into the MAGIC model by the fuzzy optimisation procedure (Jenkins *et al.*, 1997).

The expected response to the S protocol at the regional scale is recovery in the most acidified sites whilst buffered sites, which have suffered little damage in the hindcast period will show little change. Since all emissions reductions are to be completed by 2010, the stream concentration of SO_4 is constant after that time. The response of the base cations to S reductions is characterised by decreases which are more marked at the higher percentiles. There are fewer sites with high leaching of base cations in the future. The combination of anion and base cation response (Fig. 5c) is a marked increase in the 95th percentile of ANC from –49 to –20 μeq l^{-1} and, to a lesser extent, pH, such that by 2010 there are fewer sites with very high acidification status. This is consistent with mobile anion theory where the most acidified sites are expected to show the biggest recovery. Improvements in ANC are, therefore, most likely in the most acidified areas.

From 2010 to 2030, little change in base cations, ANC or pH is predicted (Figs. 4 and 5). Recovery of low ANC sites, however, continues although at a slower rate (Fig. 7). The implication is that, since anion concentrations remain constant after 2010, there is a small increase in

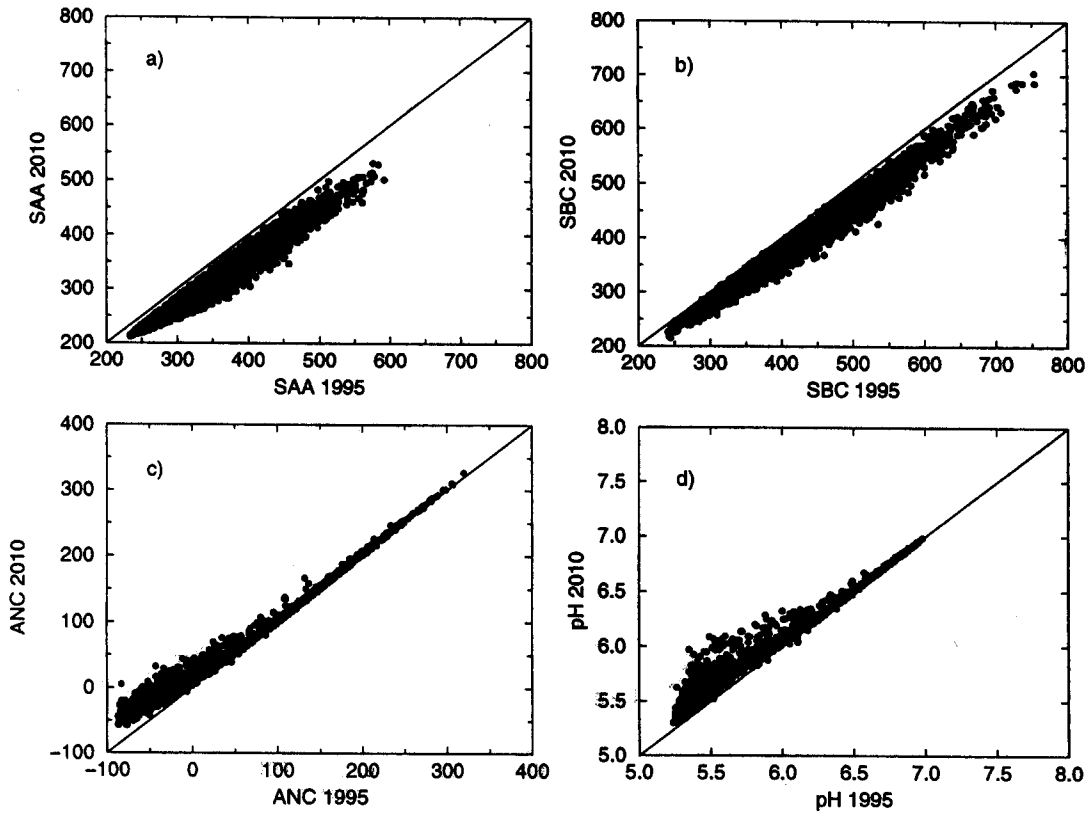

Fig. 7. *Scatter plots of simulated SAA, SBC, ANC and pH in 2010 and 2030 for 4191 acceptable simulations.*

cations as weathering replenishes depleted soil base cation stores.

A strength of the Monte-Carlo methodology is that conclusions may be drawn for Wales as a whole, since combinations of conditions not encountered in the survey, but lying within the observed ranges, are incorporated in the model. The proportion of the region with ANC <0 drops from 23% in 1984 to 19% in 1995 and further to 12% in 2010. pH and ANC in many Welsh streams are lower, however, than simulated pre-industrial levels. ANC fails to rise above zero across the entire region. The implication of these results is that further reductions in SO_4 deposition are necessary to lift ANC to levels which protect brown trout populations; the threshold adopted by the UK for national mapping of critical loads for surface waters being ANC >0 (Harriman *et al.*, 1995). Re-running the forecast with further reductions in S deposition reveal that in order to assure recovery of the 95 percentile above zero by 2030, a reduction of 70–75% in S deposition from 1989–92 to 2005, and 80–85% to 2010, was necessary. This compares with the 50% reduction resulting from the Second S Protocol.

The best and worst case HARM simulations result in low sensitivity of estimated soil and stream chemistry in 2010 and 2030. The uncertainty is greatest at the most acidified sites (Fig. 8), but is always smaller than the predicted effect of the protocol itself, since the magnitude of error in the deposition estimates is always substantially less than the estimated decrease in deposition.

Conclusions

The model simulations broadly indicate recovery of the most acidified surface waters in Wales. The recovery has already been rapid as reductions in acid deposition have occurred and is predicted to be similarly rapid as further reductions take effect. However, the Second S Protocol does not reduce emissions sufficiently for ANC in all streams in the region to rise above zero. Similar applications for the AWMN (Jenkins *et al.*, 1997) showed that at some sites, a minor recovery was followed by continued acidification as a result of soil acidification. This occurred at sites where weathering rates were insufficient to buffer continued elevated S deposition.

The decline in stream SO_4 in the regional model from 1984 to 1995 was driven by the observed decline in SO_4 deposition. The assumption has been made that the reduction in S is genuine and the lack of response in base cations in the observed data is, therefore, unusual. An alternative interpretation is that the S data in the deposition and stream surveys are unreliable, and the base cations indicate a lack of change over the 11-year period. This is unlikely

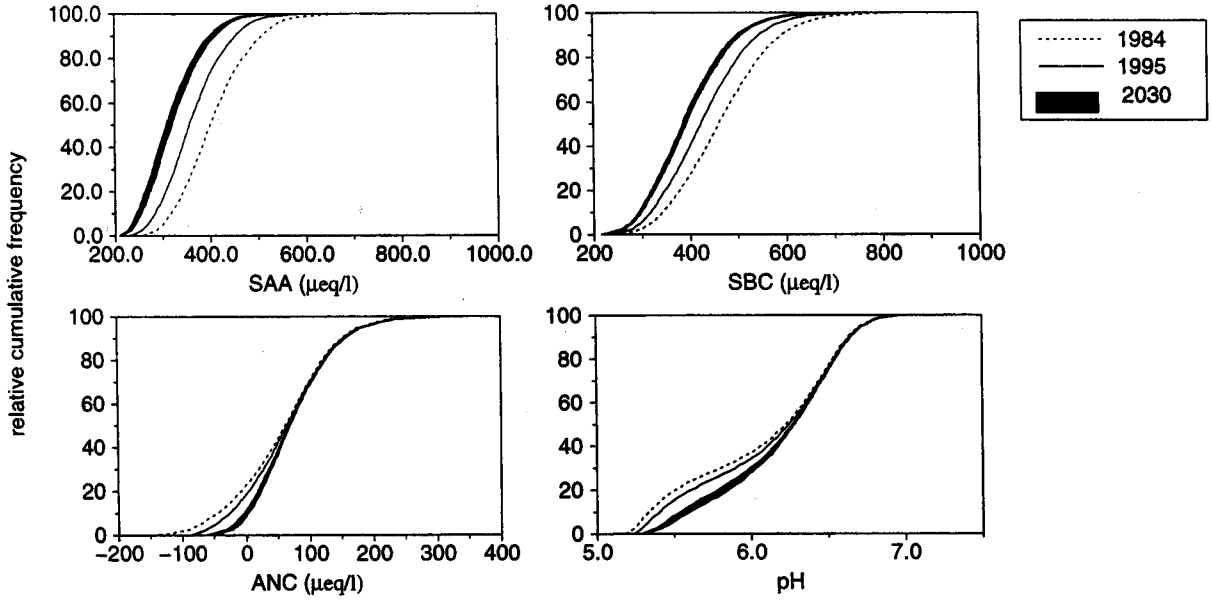

Fig. 8. *Sensitivity to uncertainty of HARM model predictions for 1984, 1995 and 2030. The band for 2030 spans the worst and best cases.*

since the observed decrease in S concentrations is consistent with observed decreases in S deposition across the UK (RGAR, 1987, 1990, 1997). Since the forecasts are based on the 1995 survey only, they are not affected by this uncertainty.

This modelling assessment incorporates no explicit modelling of land use although it is implicit in the deposition and uptake fluxes. The effects of forestry on acidification have been widely debated and are worthy of the development necessary for explicit inclusion at the regional scale. There are two possible approaches; a simplistic one would be to incorporate land use as a Monte-Carlo parameter and make some generalisations about forest age for construction of regional sequences for deposition, uptake and discharge. A more satisfactory approach is a multiple site calibration using the known land use history for each of the survey catchments (Collins and Jenkins, 1998). Furthermore, regional representation of greater complexity in the handling of N is necessary, such that mineralisation, nitrification and denitrification processes are explicitly modelled. This would also enable assessment of future N emission protocols.

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